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GENERATION OF NON-LINEAR DIPOLE MOMENT IN A TWO LEVEL SYSTEM

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ABSTRACT. With the availability of intense sources it has been possible to observe non linear effects in dipole moments in interactions where only two energy levels take part. Using the geometrical representation for Schrodinger equation the expectation value for non-linear component of dipole moment has been obtained. It has been demonstrated that a permanent dipole moment must be present in the system for generating this effect. The non-linear component is seen to be appreciable for $\omega = \omega_{12}$ and $\omega \doteq \omega_{12/2}$. The dependance of this component on the angle between the induced dipole moment and the oscillating field is shown to be $\sim$ Cos^2 θ sin θ

INTRODUCTION

The dipole moment induced in an atom, designated as a system, is seen, in general, to be linear in the frequency of the applied field. But it is possible to observe dipole moment which is not linear in the frequency of the applied field by using more intense sources than ordinarily available ones.

This generation of the higher harmonics of the frequency of an electromagnetic field interacting with a material system (or an atom) has been of much interest to physicists in recent times. (Franken and Ward, 1963). These processes, involving multiphoton transitions, may be explained by higher order perturbation theory. The field required to produce these effects must be highly intense and recently optical maser sources have been used to demonstrate this phenomena (Franken *et al.* 1961). In fact the order of magnitude of the optical maser sources to demonstrate this effect is of the order of 10^6 joules with a peak power of the order of 10^9 watts (Franken and Ward, 1963). These sources are highly monochromatic and so it is reasonable to assume that only two stationary energy states of the system (involving one transition frequency, which is more or less equal to the frequency of the applied field) will be principally involved in such an interaction. Reports are available where only two energy levels have indeed been associated in generating optical harmonics . The investigation of such multiple photon transition has been reported (Voskonyan, Klyshko and Tusmanov 1964; Klyshko and Tusmanov 1965) These investigators have used a magnetic dipole transition of the sublevels of the ground state of the free radical diphenyl picryl hydrazyl (DPPH).

The object of this article is to demonstrate the generation of harmonics of the applied field frequency in the cases where only two levels are involved in the interaction and study the characteristics of the non-linear dipole moment.

In the case of a two level system, a higher harmonic generation will be possible only if (i) there is a permanent dipole moment present in the system or (ii) if the induced dipole moment itself is non-linear in terms of the field. We shall discuss only the case (i). The fact that a permanent dipole moment is a necessity for the above mentioned processes has also been discussed in the restricted case of a magnetic dipole transition by Voskonyan, Klyshko and Tusmanov (previous reference). We shall presently demonstrate a model, which is capable of explaining the phenomenon for a more general situation.

FORMULATION OF THE PROBLEM

We start with a system of which only two non-degenerate energy levels are involved. The levels are designated as level 1 with energy E_1 and level 2 with energy E_2. Their transition frequency is ω_{12} which is always positive.

$$\omega_{12} = \frac{E_1 \sim E_2}{\hbar}$$

Only a monochromatic field is considered. The interaction coupling the system to the electromagnetic field is of electric dipole type (Senitzky 1962; Bonch) Bruevich *et al.*, 1965); the interaction hamiltonian being :

$$V(t) = -\vec{\mu} \cdot \vec{E}_0(t) \qquad \ldots \quad (2.1)$$

where $\vec{\mu}$ is the electric dipole moment operator and $\vec{E}_0$ is the applied field. The field has been regarded as classical, but a quantum theoretical consideration does not change the results (Mandel and Wolf 1963). The dipole moment operator is assumed real for convenience.

The wave function of a two level system is written down as :

$$\psi(t) = a_1(t)\psi_1 + a_2(t)\psi_2 \qquad \ldots \quad (2.2)$$

where ψ_1 is the wave function for state 1 and ψ_2 is the wave function for state 2, a_1 and a_2 being the coefficients associated with them. The phase of ψ is of little importance when only two energy levels are being considered.

The Schrodinger equation is then written down

$$(H+V)\,\psi = i\hbar \frac{d\psi}{dt} \qquad \ldots \quad (2.3)$$

H is the hamiltonian for stationary states, with the exactly solvable eigen value equations

$$H\psi_i = E_i\psi_i \qquad (i = 1, 2) \qquad \ldots \quad (2.4)$$

Substituting eqn. (2.2) into eqn. (2.3) we get

$$a_1E_1\psi_1+a_2E_2\psi_2+a_1V\psi_1+a_2V\psi_2 = i\hbar\,\frac{da_1}{dt}\,\psi_1+i\hbar\,\frac{da_2}{dt}\,\psi_2$$

Multiplying this equation with ψ_1 from left, substituting eqn. (2.4) and using the property of orthonormality of ψ_i's ($\int \psi_i^*\psi_j d\tau = \delta_{ij}$) we obtain :

$$\frac{da_1}{dt} = \frac{E_1}{i\hbar}\,a_1+\frac{V_{11}}{i\hbar}\,a_1+\frac{V_{12}}{i\hbar}\,a_2 \qquad \ldots \quad (2.5)$$

and similar equations for a_2, $a_1{}^*$ and $a_2{}^*$ can be obtained. V_{ij} is nothing but the expectation value of V and is given by $\int \psi_i^* V\psi_j d\tau$ $(i, j = 1, 2)$.

To find the expectation value of the dipole moment operator $\vec{\mu}$ directly, the geometrical representation of the Schrodinger equation (Feynman, Vernon, and Hellworth 1958) is used.

We construct the three real components of a 3-vector $\vec{r}$:

$$\begin{aligned} r_1 &= a_1a^*{}_2+a^*{}_1a_2 \\ r_2 &= i(a_1a^*{}_2-a^*{}_1a_2) \\ r_3 &= a_1a^*{}_1-a_2a^*{}_2 \end{aligned} \qquad \ldots \quad (2.6)$$

and also $r_4 = a_1a_1{}^*+a_2a^*{}_2$, which is not independant.
For a normalised ψ the value of r_4 is equal to 1 (Feynman *et al*, previous reference)

So the equation of motion for $\vec{r}$ is, using eqn. (2.5)

$$\frac{d\vec{r}}{dt} = \vec{\omega}\times\vec{r} \qquad \ldots \quad (2.7)$$

where $\vec{\omega}$ is a vector in the same hypothetical coordinate system, with

$$\begin{aligned} \omega_1 &= (V_{12}+V_{21})/\hbar \\ \omega_2 &= i(V_{12}-V_{21})/\hbar \\ \omega_3 &= \omega_{12}+(V_{11}-V_{12})/\hbar \end{aligned} \qquad \ldots \quad (2.8)$$

The expectation value of the electric dipole moment operator is directly given by :

$$\begin{aligned} \langle\vec{\mu}\rangle &= \int\psi^*\vec{\mu}\psi d\tau \\ &= \int (a^*{}_1\psi^*{}_1+a^*{}_2\psi^*{}_2)\vec{\mu}(a_1\psi_1+a_2\psi_2)d\tau \\ &= a^*{}_1a_1\int \psi^*{}_1\vec{\mu}\,\psi_1 d\tau+a^*{}_1a_2\int \psi^*{}_1\vec{\mu}\psi_2 dz+a^*{}_2a_1\int \psi^*{}_2\vec{\mu}\psi_1 d\tau \\ &\quad +a^*{}_2a_2\int \psi^*{}_2\vec{\mu}\psi_2 d\tau \\ &= \vec{\mu}_{12}r_1+\tfrac{1}{2}(\vec{\mu}_{11}-\vec{\mu}_{22})r_3+\tfrac{1}{2}(\vec{\mu}_{11}+\vec{\mu}_{22}). \end{aligned} \qquad \ldots \quad (2.9)$$

where $\vec{\mu}_{12}$ is understood to be $\int \psi^*{}_1 \vec{\mu} \psi_2 d\tau = \vec{\mu}_{21}$ when $\vec{\mu}$ is real and hermitian (Feynman *et al.*, 1958, Previous Ref.) When the field is applied the contribution to the eqn. (2.9) is predominantly due to the first term, i.e., that component of $\vec{\mu}$ which accounts for the transition between the two stationary levels.

THE SOLUTION

We assume the field to be sinusoidal ($\vec{E}_0 = \vec{E} \cos \omega t$), then the form of the interaction is :

$$\begin{aligned} V &= -\mu' \cdot E' \cos \omega t \\ &= -\mathrm{Re} \cdot \vec{\mu} \cdot \vec{E} e^{-i\omega t} \end{aligned} \qquad \ldots \quad (3.1)$$

The equation (2.7) reduces to :

$$\frac{dr_1}{dt} = -r_3 \frac{2\vec{\mu}_{12} \cdot \vec{E}}{\hbar} \sin \omega t - [\omega_{12} - (\vec{\mu}_{11} - \vec{\mu}_{22}) \cdot \vec{E}/\hbar \cos \omega t] r_2$$

$$\frac{dr_2}{dt} = [\omega_{12} - (\vec{\mu}_{11} - \vec{\mu}_{22}) \cdot \vec{E}/\hbar \cos \omega t] r_1 + r_3 \frac{2\mu_{12} \cdot E}{\hbar} \cos \omega t$$

$$\frac{dr_3}{dt} = r_2 \frac{2\vec{\mu}_{12} \cdot \vec{E}}{\hbar} \cos \omega t - r_1 \frac{2\vec{\mu}_{12} \cdot \vec{E}}{\hbar} \sin \omega t \qquad \ldots \quad (3.2)$$

If one neglects $\mu_{11}/\hbar$ and $\mu_{22}/\hbar$ in comparison with ω_{12}, equation (3.2) are formally similar to Bloch equations of magnetic susceptibility (Bloch 1946), but are more general. and can be solved exactly by the rotating coordinate method, (Archibald 1952) to find the value of linear dipole moment. But the presence of a time dependance in ω_3 component complicates the situation.

The rotating coordinate transformation

$$\begin{aligned} r'_1 &= r_1 \cos \omega t + r_2 \sin \omega t \\ r'_2 &= -(r_1 \sin \omega t - r_2 \cos \omega t). \\ r'_3 &= r_3 \end{aligned} \qquad \ldots \quad (3.3)$$

with its inverse

$$\begin{aligned} r_1 &= r'_1 \cos \omega t - r_2 \sin \omega t \\ r_2 &= r'_1 \sin \omega t - r'_2 \cos \omega t \\ r_3 &= r'_3 \end{aligned} \qquad (3.4)$$

gives us the equations :

$$\frac{dr'}{dt} = \omega' \times r' \qquad \ldots \quad (3.5)$$

where

$$\omega'_1 = \gamma = -\frac{2\vec{\mu}_{12}\cdot\vec{E}}{h}$$

$$\omega'_2 = 0$$

$$\omega'_3 = \Delta+\delta\cos\omega t \text{ with } \Delta = \omega_{12}-\omega;\ \delta = -(\vec{\mu}_{11}-\vec{\mu}_{22})\cdot\vec{E}/\hbar$$

We proceed by writing for the $\vec{r}'$ components :

$$\begin{aligned} dr_1'/dt+[\Delta+\delta\cos\omega t]r_2' &= 0 \\ dr_2'dt-[\Delta+\delta\cos\omega t]r_1+\gamma r_3 &= 0 \\ dr_3'/dt-\gamma r_2' &= 0 \end{aligned} \quad \dots \quad (3.6)$$

This set of equations is not exactly solvable, but it is possible to solve them by an approximation.

To solve the equations (3.6) the following substitutions are made .

$$\begin{aligned} r_1' &= \sum_{n=-\infty}^{+\infty} u_n(\gamma,\delta)e^{-in\omega t} \\ r_2' &= \sum_{n=-\infty}^{+\infty} v_n(\gamma,\delta)e^{-in\omega t} \\ r_3' &= \sum_{n=-\infty}^{+\infty} r_n(\gamma,\delta)e^{-in\omega t} \end{aligned} \quad \dots \quad (3.7)$$

It is easy to see that $(n = 0)$ terms are nothing but the contributions to r_1 and r_2 which are linear in the applied field, whereas $(n = 0)$ term in r_3 is non-oscillatory.

Substituting the equation (3.7) into equation (3.6) one gets :

$$\begin{aligned} -\frac{in\omega}{\gamma}u_n+\frac{\Delta}{r}v_n+\frac{1}{2}\frac{\delta}{\gamma}(v_{n-1}+v_{n+1}) &= 0 \\ -\frac{in\,\omega}{\gamma}v_n-\frac{\Delta}{\gamma}u_n-\frac{1}{2}\frac{\delta}{\gamma}(u_{n-1}+u_{n+1})+r_n &= 0 \\ -\frac{in\,\omega}{\gamma}r_n-v_n &= 0 \end{aligned} \quad \dots \quad (3.8)$$

We substitute $q = \frac{\delta}{\gamma}$ and $s = \frac{\Delta}{\gamma}$ and assume $q < <1$. This assumption is quite justified as the necessity of having a highly intense field to observe the non linear effects points out that δ is indeed very much smaller that γ. We now expand u_n, v_n and r_n in powers of q to see how these terms depend on this parameter

$$u_n(q) = \sum_{\nu=0}^{\infty} u_{n\nu} q^\nu$$

$$v_n(q) = \sum_{\nu=0}^{\infty} v_{n\nu} q^\nu \qquad \dots \quad (3.9)$$

$$r_n(q) = \sum^{\infty} r_{n\nu} q^\nu$$

These equations allow us to write equations (3.8), after picking up similar coefficients of q, in the form :

$$-\frac{in\omega}{\gamma} u_{n\nu} + s v_{n\nu} + \frac{1}{2}(v_{n-1,\,\nu-1} + v_{n+1,\,\nu-1}) = 0$$

$$-\frac{in\omega}{\gamma} v_{n\nu} - s u_{n\nu} - \tfrac{1}{2}(u_{n-1,\,\nu-1} + u_{n+1,\,\nu-1}) + r_{n\nu} = 0 \qquad \dots \quad (3.10)$$

$$-\frac{in\omega}{\gamma} r_{n\nu} - v_{n\nu} = 0$$

We shall also consider the symmetry properties of u_n, v_n and r_n, which are :

$$u_n(-q) = (-1)^n u_n(q)$$
$$v_n(-q) = (-1)^n v_n(q) \qquad \dots \quad (3.11)$$

and

$$r_n(-q) = (-1)^n r_n(q)$$

which means in equations (3.9) only even terms of the combination $n+\nu$ will exist. An additional fact must also be considered : when q is zero, r_n cannot have an oscillatory term dependant on ω, as under such a circumstance the equations (3.6) can be shown to be exactly solvable. So the terms like r_{20}, r_{40} are nonexistent.

$$r_{n0} = r_{n0}\delta_{n0} \qquad \dots \quad (3.12)$$

Now we shall solve equations (3.10) for specific n and ν.

For $\nu = 0$, the equations are independant of q, and reduce to the simpler case where there is no permanent dipole moment. So the equations reduce to :

$$-\frac{in\omega}{\gamma} u_{n0} + s v_{n0} = 0$$

$$-\frac{in\omega}{\gamma} v_{n0} - s u_{n0} + r_{n0} = 0 \qquad \dots \quad (3.13)$$

$$-\frac{\omega\, ni}{\gamma} r_{n0} - v_{n0} = 0$$

It can be clearly seen, using equation (3.12) that for $n \neq 0$,

$$u_{n0} = u_{n0}\delta_{n0} \quad \text{and} \quad v_{n0} = v_{n0}\delta_{n0}$$

which means no higher order solution will exist unless there is a permanent dipole moment present $(\delta \neq 0)$.

THE NON LINEAR DIPOLE MOMENT

The first order solutions of u_n and v_n with $n = 1$ give rise to $u_{n\nu}$ and $v_{n\nu}$ values which give a contribution to the value of dipole moment which oscillates with a frequency 2ω.

The same approximation also gives a contribution to the linear part of the dipole moment expectation value $< \vec{\mu} >$ through $r_{n\nu}$.

Writing equations (3.10) for $n = 1$, $\nu = 1$

$$
\begin{aligned}
&-\frac{i\omega}{\gamma}\, u_{11} + s v_{11} + \frac{1}{2}\, r_{00} = 0 \\
&-\frac{i\omega}{\gamma}\, v_{11} - s u_{11} - \frac{1}{2}\, u_{00} + r_{11} = 0 \qquad \ldots \quad (4.1) \\
&-\frac{i\omega}{\gamma}\, r_{11} - v_{11} = 0
\end{aligned}
$$

We substitute the value of u_{00} and v_{00} from equations (3.13) ($u_{00} = \gamma/\Delta r_{00}$ $v = 0$) and get :

$$u_{11} = \frac{\gamma^2}{2(\omega^2 - \Delta^2 - \gamma^2)}\, r_{00}$$

$$v_{11} = \frac{i\omega\gamma^2}{2\Delta(\omega^2 - \Delta^2 - \gamma^2)}\, r_{00} \qquad \ldots \quad (4.2)$$

These two terms give the major contribution to the part of dipole moment which has a frequency of 2ω. The value of r_{11} may also be noted and this will give a correction to the expectation value of dipole moment with a frequency of ω :

$$r_{11} = -\frac{\gamma^3}{2\Delta(\omega^2 - \Delta^2 - \gamma^2)}\, r_{00} \qquad \ldots \quad (4.3)$$

Similarly we may carry on the higher order calculations, and get 3ω, 4ω, ... dependant values of dipole moment expectation value.

Now collecting the u_{11}, v_{11} values and tracing back the substitutions

$$
\begin{aligned}
r_1' &= u_{00}+u_{11}qe^{-i\omega t}+\ldots \\
r_2' &= v_{11}qe^{-i\omega t}+\ldots \\
r_3' &= r_{00}+r_{11}qe^{-i\omega t}
\end{aligned} \qquad \ldots (4.4)
$$

Collecting only the real values and operating the inverse transformation eqn. (3.4)

$$
r_{1_{dc}\atop 2\omega} = \frac{\gamma\delta}{4(\omega^2-\Delta^2-\gamma^2)}\left[1-\frac{\omega}{\Delta}\right]r_{00}+\frac{\gamma\delta}{4(\omega^2-\Delta^2-\gamma^2)}\left[1+\frac{\omega}{\Delta}\right]r_{00}\cos 2\omega t
$$

$$
r_{2_{2\omega}} = \frac{\gamma\delta}{4(\omega^2-\Delta^2-\gamma^2)}\left[1-\frac{\omega}{\Delta}\right]r_{00}\sin 2\omega t \qquad \ldots (4.5)
$$

DISCUSSION

The equations (4.5) give us the value of non linear dipole moment, both the non oscillatory part and the part which has a dependance on 2ω. The non oscillatory contribution to the expectation value, using eqn. (2.9) is

$$
\langle\vec{\mu}\rangle_{dc} = \frac{\gamma\cdot\delta\vec{\mu}_{12}(\omega_{12}-2\omega)}{4(\omega_{12}-\omega)(\omega^2-\Delta^2-\gamma^2)}\,r_{00}+\tfrac{1}{2}(\vec{\mu}_{11}-\vec{\mu}_{22})r_{00}+\tfrac{1}{2}(\vec{\mu}_{11}+\vec{\mu}_{22}) \qquad (5.1)
$$

The 2ω dependant part is, neglecting the second order term in r_3 :

$$
\langle\vec{\mu}\rangle_{2\omega} = \frac{\gamma\cdot\delta\vec{\mu}_{12}\omega_{12}}{4(\omega_{12}-\omega)[\omega^2-(\omega_{12}-\omega)^2-\gamma^2]}\,r_{00}\cos 2\omega t \qquad \ldots (5.2)
$$

As we are dealing with high frequencies, one can neglect γ^2 in comparison with ω^2 in equations (5.1) and (5.2). This only means that field saturation is not considered. Then equations (5.1) and (5.2) reduce to :

$$
\langle\vec{\mu}\rangle_{dc} = -\left[\frac{\gamma\delta\vec{\mu}_{12}}{4(\omega_{12}-\omega)\omega_{12}}-\tfrac{1}{2}(\vec{\mu}_{11}-\vec{\mu}_{22})\right]r_{00}+\tfrac{1}{2}(\vec{\mu}_{11}+\vec{\mu}_{22})
$$

$$
\langle\vec{\mu}\rangle_{2\omega} = \frac{\gamma\delta\vec{\mu}_{12}}{4(\omega_{12}-\omega)(2\omega-\omega_{12})}\,r_{00}\cos 2\omega t \qquad \ldots (5.3)
$$

In general, for a complex value of dipole moment, the value of $\langle\vec{\mu}\rangle$ must contain r_2 contribution, which is :

$$
\langle\mu\rangle_{2\omega} = \frac{\gamma\delta\,\mu_{12}}{4(\omega_{12}-\omega)\omega_{12}}\,r_{00}\sin 2\omega t \qquad \ldots (5.4)
$$

These two equations, eqn. (5.3) and (5.4), show an interesting property of the 2ω dependant part of $<\vec{\mu}>$ that is, $<\vec{\mu}>_{2\omega}$. One gets the maximum value for $<\vec{\mu}>_{\omega}$ for two values of ω : $\omega = \omega_{12}$ and $\omega = \omega_{12}/_2$. In the second case the value of $<\vec{\mu}>_{\omega}$ has already fallen off, so that at $\omega_{12} = 2\omega$ the effect of $<\vec{\mu}>_{\omega 2}$ will be easily observed. Both the effects-giving rise to an absorption of second harmonic power can be in principle observed.

The equations (5.3) and (5.4) do not immediately display saturation due to finite width of energy levels. But as pointed out by Haaken *et al.* (Haaken, der Agobian and Pauthier 1965), one can directly take into account this effect by replacing ω_{12} by a complex quantity $\omega_{12}'+i/\Gamma$; where Γ has the significance of a relaxation time. The equations (5.3) and (5.4) also point out that the orientation of the oscillating field affects the value of $<\vec{\mu}>_{2\omega}$ in a very different manner than the value of $<\vec{\mu}>_{\omega}$. $<\vec{\mu}>_{\omega}$ directly depends on $\vec{\mu}_{12}\cdot\vec{E}$, so that if the angle between $\vec{\mu}_{12}$ and $\vec{E}$ is Θ; then the angular dependance of $<\vec{\mu}>_{\omega}$ is

$$<\mu>_{\omega} \sim \cos^2\Theta$$

when we consider it in the direction of the applied field. It will show a maximum at $\Theta = 0$ and drop off to zero at $\Theta = \pi/2$. But the dependance of $<\vec{\mu}>_{2\omega}$ is as follows :

$$<\vec{\mu}>_{2\omega} \sim (\vec{\mu}_{12}\cdot\vec{E})[(\vec{\mu}_{11}-\vec{\mu}_{22})\cdot\vec{E}](\vec{\mu}_{12}\cdot\hat{E})$$

$\hat{E}$ is the unit vector in the direction of $\vec{E}$.

If one uses a static (non oscillating) field to separate the levels (as the strong static magnetic field in a Bloch system) $\vec{\mu}_{12}$ will be in the direction of this field and $(\vec{\mu}_{11}-\vec{\mu}_{22})$ will be perpendicular to it. This is evident from the fact that the transition inducing dipole moment may be denoted by Pauli σ_x, σ_y matrices and the permanent dipole moment may then be denoted by the perpendicular σ_z matrix, as pointed out by Dicke (Dicke 1954). Then the angular dependance of $<\vec{\mu}>_{2\omega}$ is:

$$<\mu>_2 \qquad \Theta \sin\Theta$$

This is, of course, strictly true for the part of $<\vec{\mu}>_{2\omega}$ when $\omega = \omega_{12/2}$. In the case $\omega = \omega_{12}$; r_2 value has to be considered and the expression is expected to be more involved.

The expression for $<\vec{\mu}>_{2\omega}$, where $2\omega = \omega_{12}$, now shows that the maximum does not occur at $\Theta = 0$. In fact $<\vec{\mu}>_{2\omega}$ is equal to zero for $\theta = \pi/2$ and

$\Theta = 0$. The maximum occurs at about $\Theta \sim 35°$. Such a result has been verified in the case of a magnetic dipole transition by Voskonyan *et al.* (Voskonyan *et al.* 1964).

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